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ABSTRACT

A standardized, reproducible method of surrogate dermal monitoring was devised to supplement knowledge of the potential transfer of pesticide residues from floor surfaces to persons in contact with the floor. This device was a 12 kg. foam-covered rolling cylinder equipped with stationary handles. The device was rolled over a cotton cloth (the actual collection media) placed over carpet to be sampled. This method transfers between 1 and 3 percent of the potential available pesticide material from nylon carpeting to the collection media. Transfer from carpet to cotton cloth correlates highly with transfer to cotton clothing worn by persons exercising on the carpet.

INTRODUCTION

The use of home pesticide foggers results in estimated deposition on the floor of 55 to 69 percent (Ross et al. 1990) of its contents. Floors typically contain a majority of the non-volatile fogger contents (Maddy et al. 1987), resulting in a considerable potential for dermal transference to a person in contact with treated carpet. A recent study (Ross et al. 1990) investigated the transfer of two pesticides from a carpeted floor to dosimetery clothing worn by adults exercising on the treated floor. However, the use of human subjects may not always be possible because of questions of subject safety; the need to test a variety of formulations, pesticide products, floor coverings or furnishings; constraints on facilities and/or funding. Therefore, in conjunction with the human exposure study, a parallel study of the effectiveness of a surrogate dislodgement device (carpet roller) was performed.

METHODS AND MATERIALS

Several identical 7.5 oz home-fogger devices, (K-RID^R Brand, K-Mart Stores distributors; Chemsico Company, manufacturer; EPA Reg.# 9688-63) which contain chlorpyrifos as the predominate active ingredient, were purchased in a local retail store. All chlorpyrifos-containing foggers sold in California are formulated and packaged by one company (Chemisco, St. Louis, MO). Two unopened foggers were sent to the California Department of Food and Agriculture's (CDFA) Chemistry Laboratory Services for analysis of chlorpyrifos and d-trans allethrin. Analysis confirmed the presence of the two active ingredients at the levels indicated on the label (0.5 percent for chlorpyrifos, 0.05 percent for d-tran allethrin).

One fogger was discharged in each of eight test rooms, according to label directions. These test rooms were located in a large hotel in Sacramento, California. Details of the room dimensions, configurations and environmental conditions are provided in Ross, et al. The facility floors were untreated, 100% nylon cut-pile carpet.

Carpet swatches of material not the same type as the facility carpet were layed out on the floor of two rooms, (designated Physicochemical Rooms in the Ross et al. study) forming a triad with arms radiating 120° from the fogger. Carpet swatches were of two grades/types: low-cost 100% nylon carpeting and a higher, denser pile 100% nylon carpeting that had been made with a stain resisting material known as TREVIRA^{R}. Swatches' area were 1450 cm^2. Stain resistant carpet was placed closer to the fogger than the less dense swatches.

Absorbent cotton gauze dosimeters (exposed area of 23.76 cm^2) were distributed in all rooms. These were also used for comparison of the ability of the roller method to effect pesticide transfer from the floor to the cotton roller sheet. Details of the protocol for the gauze dosimeters are contained in Ross et al. (1990).

The carpet roller was constructed as indicated in Figure One, using the following procedure: Drill 0.5" hole in center of an ABS sewer pipe cap (exterior covering, 4" diameter). Slide 4" PVC pipe (0.75" diameter) over quarter threaded 6" bolt (0.5" diameter). Slide one washer (0.5" diameter center hole) washer over bolt section projecting from PVC pipe. Slide this handle assembly into the hole on the convex side of the ABS sewer cap. Slide another duplicate washer onto bolt where it projects into the concavity of the ABS cap. Screw matching hexnut (0.5" inner diameter) onto the bolt and tighten, securing the handle assembly onto the cap. Repeat with a second cap/handle assembly. Cap one end of a 2' long by 4" diameter sewer pipe with the cap/handle assembly. Fill pipe with lead shot ballast (approximately 25 pounds). Cap other end. Optionally, secure caps with duct tape or plastic adhesive. Cut one sheet of 1/2" high density polyurethane foam to fit on pipe between ridges formed by cap ends. Use enough foam to wrap around the pipe and raise the center pipe portion (the part in contact with the floor) higher than the cap ridges. Secure the foam with duct tape.

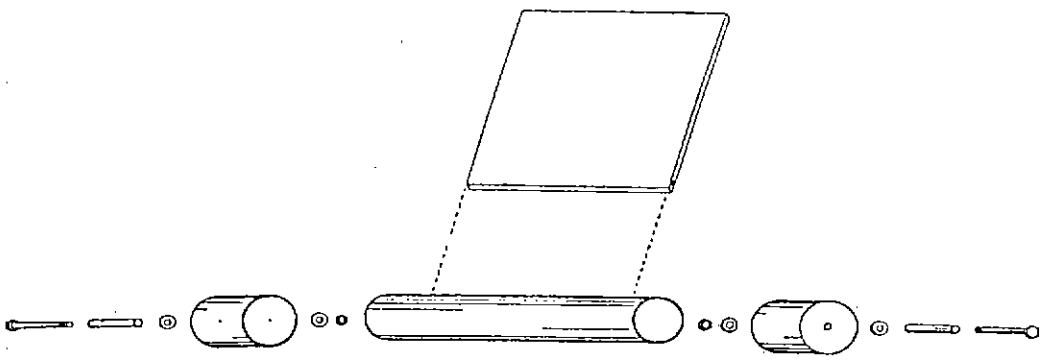

Figure One
Schematic view of CDFA Carpet Roller

After venting the rooms according to the label following fogger use, carpet swatches were collected and placed outside their test rooms. A sheet of cloth made from percale bedsheets (50% cotton/ 50% Kodel^R polyester, 180 thread count, 1840 cm^2 $\pm$90 cm^2) was placed over each carpet swatch. A sheet of plastic (GLAD^R Brand Tall Trash Bag, white) was placed over the cloth sheet. The roller was rolled over the plastic/cloth/carpet sandwich ten times. One forward push and one backward pull constituted one roll. Care was taken not to press down on the roller which would have added extra weight. After rolling, the plastic was discarded and the cloth was carefully removed from the carpet and placed into a wide-mouth quart MASON^R jar. The jar was sealed with aluminum foil, capped, and placed on dry ice.

In the rooms in which human subjects exercised to estimate potential dermal transfer from the carpet, two areas not contacted by the test subjects were identified. The times of each sampling were 0, 6 and 12.5 hours post application (2 rooms per time period). Immediately following the Jazzercise^R portion of the test, one percale cloth was placed on each non-contact area. The cloths were layed out flat on the facility carpet. Each cloth was located in opposite corners from each other (far left and far right with respect to the entry way). A plastic sheet was placed over the cloth and this assembly was rolled 10 times, as described earlier. The cloths were then taken off the floor, placed in separate plastic bags (ZIP-LOC^R brand bags, MASON^R jars being unavailable during this phase) and stored on dry ice for later analysis. Additionally, in the rooms that were sampled at 0 hours post-venting, two other sampling strategies were employed. One was a resampling of facility carpeting, on the same place sampled earlier, to measure the efficacy of the roller. The second was a sampling from the area under one of the test subjects to investigate the amount of material removed by the activity of the dosimeter-clad subjects.

Analysis was performed by CDFA Chemistry Laboratory Services. Analysis was done for chlorpyrifos, its oxon and d-trans allethrin.

RESULTS

Transferrability results are shown in Table One for carpet swatches.

TABLE ONE: Transferred residue values (in ug/cm^2) from dosimeter carpet material ("swatches") to percale cloth using the CDFA carpet roller device. Roman numerals identify replicate rooms.

	Stain Resistant Carpeting			Standard Nylon Carpeting		
Chlopyrifos I	0.07	0.19	0.61	0.06	0.13	0.12
Chlopyrifos II	0.14	0.11	0.08	0.08	0.06	0.08
	MEAN = 0.20 $\pm$0.21			MEAN = 0.09 $\pm$0.03		
d-trans Allethrin I	0.02	0.01	0.01	0.01	0.01	0.01
d-trans Allethrin II	0.01	0.03	0.07	0.01	0.01	0.01
	MEAN = 0.025 $\pm$0.024			MEAN = 0.01		

Because of the closer proximity of the stain-resistant carpet to the fogger, it is not clear whether increased removal from stain-resistant carpet is due to increased deposition or decreased permeability/absorption of carpet fibers.

TABLE TWO: Transferred residue values (in ug/cm^2) from facility carpet material to percale using the CDFA carpet roller device. Mean gauze dosimeter (MGD) values also presented. Roman numerals identify replicate rooms.

FACILITY CARPET ROLLER RESULTS

	Chlorpyrifos	d-trans Allethrin
Zero Hours Post-Application (Immediate Post)		
Right quadrant I	0.048	0.0055
Right quadrant II	0.106	0.0124
Left quadrant I	0.040	0.0048
Left quadrant II	0.027	0.0028
MEAN	0.055 ±0.035	0.0064 ±0.0042
MGD	2.36	0.2175
Six Hours Post-Application		
Right quadrant I	0.058	0.0104
Right quadrant II	0.022	0.0045
Left quadrant I	0.015	0.0031
Left quadrant II	0.026	0.0061
MEAN	0.030 ±0.019	0.0060 ±0.0032
MGD	2.311	0.2350[b]
Twelve and one-half Hours Post-Application		
Right quadrant I	0.048	0.0087
Right quadrant II	0.016	0.0033
Left quadrant I	0.013	MDL
Left quadrant II	0.014	MDL
MEAN	0.023 ±0.017	0.0044 ±0.0029[c]
MGD	2.019	0.2450

[a]MDL - Minimum Detectable Value (chlorpyrifos = 0.0005, d-trans = 0.0027)

[b]Derived from different room series (Physicochemical vs. Jazzercise[R] Exposure Room) gauze data since no gauze d-trans allethrin samples were taken in the appropriate room by Ross et al., 1990.

[c]Includes MDL values from left quadrants.

No chlopyrifos oxon was detected on any carpet roller samples. Unexposed control rooms had no detectable levels of d-trans allethrin and two samples (of four) which were 2X above the MDL (0.001 ug/cm^2) for chlorpyrifos.

The carpet roller method for transfer sampling appears to transfer approximately 1 to 3 percent of the floor residue, when comparing mean gauze pad residues to the amount of material transferred to the roller sheet. Results are shown graphically in Figure 2. The transferrability of both chlorpyrifos and d-trans allethrin declines, with a half-life of ~10 hours and ~20 hours, respectively, over the 12 hour test period.

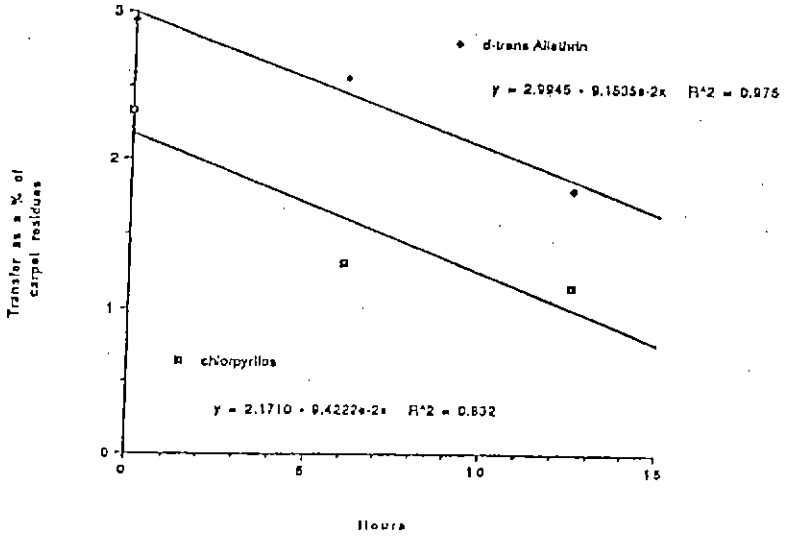

Figure Two
Time-dependant reduction in transfer of fogger residues as measured using CDFA roller.

The immediate post-application areas that were resampled by rerolling, recovered 69 percent of the first mean roller recovery value for chlorpyrifos and 75 percent of the first mean roller recovery for d-trans allethrin. Results are shown in Table Three.

TABLE THREE: Transference of disturbed (post-exercise tested rooms) pesticide residues immediately post-venting (in ug/cm^2).

	Chlorpyrifos	d-trans Allethrin
Right quadrant I	0.025	0.0027
Right quadrant II	0.078	0.0098
Left quadrant I	0.028	0.0038
Left quadrant II	0.020	0.0027
MEAN	0.038 $\pm$0.027	0.0048 $\pm$0.0034

These values are less than amounts transfered prior to human dosimeter contact, thought not statistically significant at $p<0.05$ by Students t-test. This suggests that the reservoir of transferable pesticide residue is limited and decreases following initial contact so that subsequent contacts will not transfer nearly as much as the initial contact.

Roller samples were also taken from the area that had been used by one of the dosimeter test subjects. Two samples (roll and re-roll) were taken in each of the immediate-post-venting rooms.

TABLE FOUR: Results of sampling areas directly under one of the dosimeter test subjects, using the carpet roller. Samples were taken twice from same carpet area with minimal time between samples. Values are in ug/cm^2.

	Chlorpyrifos	d-trans Allethrin
Under Subject I	0.037	0.0046
Under Subject I^{rr}	0.017	0.0031
Under Subject II	0.020	0.0031
Under Subject II^{rr}	0.015	0.0027

^{rr}re-rolled (two samplings from exact same area).

When the sequential transfers are compared (zero-hour post-venting carpet roller residue versus under subject residue versus under subject residue reroll) a pattern becomes evident. The mean zero-hour chlorpyrifos residue was 0.055 ug/cm^2. The mean value for the under subject residue was 0.029 ug/cm^2. The mean value for the under-subject rerolls was 0.016 ug/cm^2. Table Five shows the results of comparing the three residue levels. As Table Five shows, the mean percent difference from either removing residue by dosimeter clothing in contact with carpet or by the carpet roller system is similar for chlorpyrifos (47% vs. 45%). This suggests that the carpet roller may be an exceptionally good surrogate for human exposure studies for chlorpyrifos. The sequential transfer loss of d-trans allethrin is also quite comparable both after exercise and after reroll.

TABLE FIVE: Sequential comparison of mean carpet pesticide residue levels before contact with human dosimeter subject; after contact and after a second rerolling on the same carpet area. All residue values in ug/cm^2.

Before Contact Value	After Contact Value	Percent Difference	After Contact Value	Reroll Value	Percent Difference
		CHLORPYRIFOS			
0.055	0.029	-47%	0.029	0.016	-45%
		ALLETHRIN			
0.0064	0.0039	-39%	0.0039	0.0029	-26%

Residues on the dosimeter clothing (in ug of pesticide per cm^2 of body surface) of individuals exercising on a chlorpyrifos/d-trans allethrin treated floor (Ross et al.) can be graphed as a function of the residues transferred to the percale cloth by the CDFA roller (Figures Three and Four). The high regression correlation coefficients indicate that the

roller may be an excellent predictive tool for indoor exposure assessment. Each time point represents a different "aged" carpet residue being transferred at a constant rate to both percale cotton and cotton dosimeter clothing.

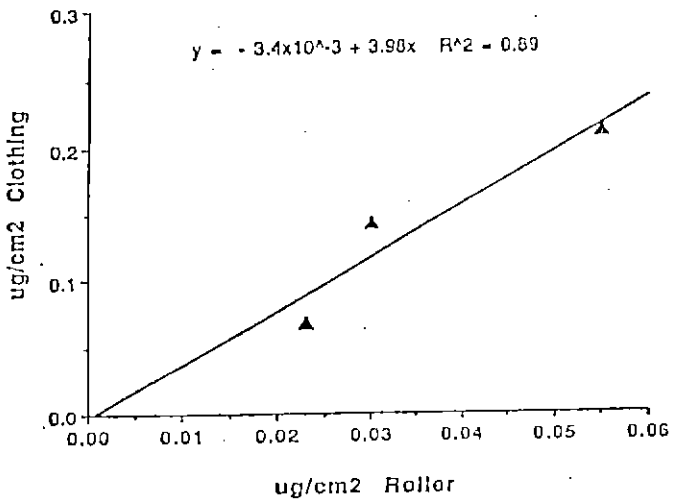

Figure Three
Correlation of Residue Transfer
Roller vs. Clothing
Chlorpyrifos

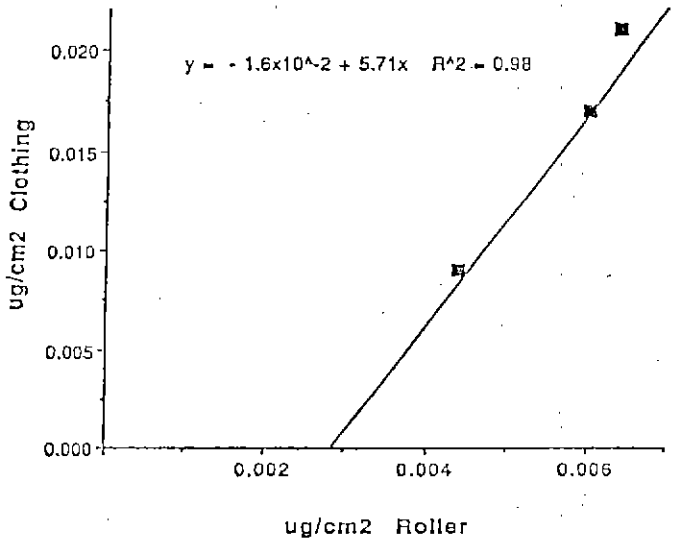

Figure Four
Correlation of Residue Transfer
Roller vs. Clothing
d-trans Allethrin

Using a procedure developed for agricultural pesticide monitoring to estimate dermal exposure based on dislodgeable foliar residue (Zweig, et al., 1985), transfer factors for dermal exposure, based on residue collected by the roller, can be calculated. The equation used:

$$\frac{\text{(total ug residue on clothing ÷ time of exposure in hours)}}{\text{ug/cm}^2 \text{ residue on roller sheets}}$$

results in a transfer factor value expressed in cm^2 contacted by the person per hour. The human dosimeter values from Ross, et al. were generated from the same rooms as the roller sheets and can be used for this computation. The exposure time was 20 minutes (0.333 hours). Applying the mean residue values to the equation results in mean transfer coefficients of 200,000 ±50,000 cm^2/hour for chlorpyrifos and 140,000 ±30,000 cm^2/hour for d-trans allethrin.

DISCUSSION AND CONCLUSIONS

Figure Two indicates that both pesticides are transfered linearly over time when analyzed as a function of time versus percent transfer. Although the two lines are parallel, the pesticides are not transfered at the same rate. Additionally, as indicated in Table Three, the reservoir of transferable pesticide residue is rapidly limited and decreases following initial contact so that subsequent contacts will not transfer nearly as much as the initial contact. The decline in transfer may reflect several possible factors:

1. Loss of solvent inerts (via evaporation/absorption/adsorption) which maintain the pesticide in a transferable thin film solution.

2. Absorption of the pesticide into the carpet fiber.

3. Irreversable absorption (chemical or electrostatic binding) of the pesticide onto the carpet fiber surface.

4. Degradation into non-detected/detectable products.

5. Volatilization of the pesticide into the atmosphere.

6. Migration of the pesticide, either independently or attached to a dust particile, into areas not available for contact by the carpet roller (e.g. carpet backing or foam pad).

Figure Five illustrates the the physical redistribution processes contained in the abovementioned points 2, 3, 5, and 6.

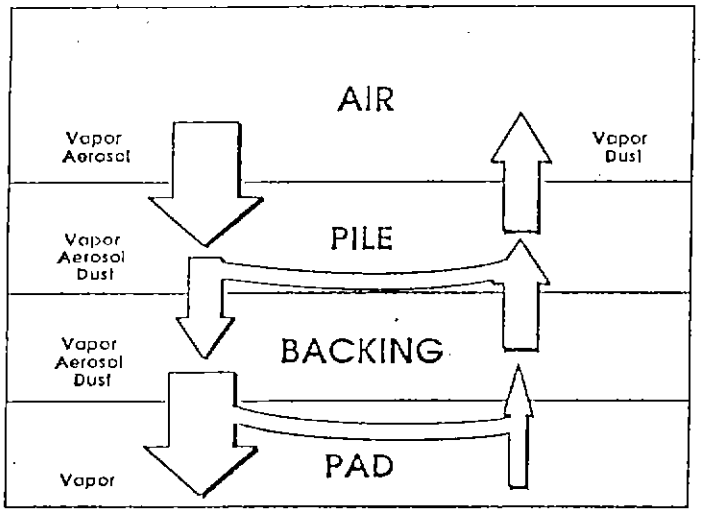

Figure Five
Redistribution of Pesticides Applied to Carpet

Recently a polyurethane foam (PUF) cylinder on a stainless steel roller was employed to investigate the sampling differences between that roller and a hand pressing technique for transfer of pesticides on aluminum foil (Hsu, et al. 1990). The pressure exerted by the PUF roller, a hand pressing method, a child (1 year old, 20 pounds weight) crawling and walking were calculated. These were 7.4 kPa, 6.9 kPa, 6.9 kPa and 8.6 kPa, respectively. The CDFA roller exerts a force of 5.6 kPa. The CDFA roller appears to transfer proportional amounts of pesticide compared to transfers measured by human dosimetry (Ross et al., 1990). The primary advantage of the CDFA roller appears to be the ease of replacement of the residue

sampling media which consists of a percale sheet. The PUF roller requires a time-consuming removal and replacement of the foam cylinder which necessitates increased sample handling and potential contamination/transfer loss.

Other methods have been devised to monitor surface pesticide residues. The wipe method (Fenske et al., 1990; Health and Welfare Canada, 1989; Cal/OSHA, 1990) has long been used as a way to sample surface residue, both pesticidal and other chemicals. The drag method (Vacarro, 1990) has also been used to monitor chlorpyrifos carpet residues.

Both the wipe method and the drag method may have drawbacks not experienced with a roller method. The wipe method's greatest deficiency is its lack of reproduceible pressure exerted by different (or even the same) investigators. This non-reproducibility may result in sample data of low precision. The drag method, though unlike the wipe in that the pressure exerted can be made constant, has a velocity component that makes reproducibility somewhat difficult. If investigators pull the drag at different speeds, this may affect the interaction of the sampling media and the sampled surface, especially on rough, uneven carpet surfaces.

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